APPLICATIONS OF RECENT DEVELOPMENTS IN X-RAY EQUIPMENT AND TECHNIQUES

BY

WILLIAM FREDERICK WAGNER

A. B., Culver-Stockton College, 1938

M. S., University of Chicago, 1940

AN ABSTRACT OF A THESIS

SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY IN CHEMISTRY IN THE GRADUATE SCHOOL OF THE UNIVERSITY OF ILLINOIS, 1947

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APPLICATIONS OF RECENT DEVELOPMENTS IN X-RAY EQUIPMENT AND TECHNIQUES

Introduction

With the phenomenal growth in the use of X-ray methods in many fields has come the inevitable improvement of X-ray equipment and techniques. One recent development from years of research in this laboratory was the Machlett Model AEG-50 X-ray tube by Rogers (1) in response to the need for a much higher intensity of radiation in studying bactericidal and photochemical effects of X-rays. This tube, operating at 50 KV and 50 MA, gives a dosage of approximately 2,500,000 roentgen units per minute at the window. Another recent innovation was the Norelco Geiger-Counter X-ray Spectrometer by Friedman (2) designed primarily for taking X-ray diffraction patterns of powdered samples. A third development was the application of a monochromatic X-ray source for locating small defects in aluminum castings by Clark and Eyler (3).

The purpose of this investigation was: first, to devise a method of chemical analysis by X-ray spectroscopy using the Machlett AEG-50 tube for a powerful primary X-ray source with the extremely sensitive Geiger-Counter Spectrometer for analysis of the secondary radiation from the sample; second, to study the antigenicity of *Eberthella typhosa* killed by radiation from the high intensity tube; and third, to extend the use of monochromatic radiation to medical radiography and therapy.

PART I

CHEMICAL ANALYSIS BY SECONDARY X-RAY EMISSION SPECTRA USING A HIGH INTENSITY X-RAY SOURCE AND A GEIGER COUNTER SPECTROMETER

Introduction

Methods used in X-ray spectroscopy and its applications to chemical analysis are thoroughly discussed by Siegbahn (4) and von Hevesy (5). Clark (6) has made a concise summary of the apparatus used, advantages and disadvantages of the methods used in X-ray spectrographic analysis. The two most commonly used methods are: (1) Measurement of primary spectral emission lines in which the material undergoing analysis is made the target of an X-ray tube. (2) Measurement of the secondary fluorescent emission lines, in which the unknown is so placed on some device inside the X-ray tube that it is screened from the cathode rays, but directly irradiated by the primary beam. These methods that require highly specialized equipment and skilled operators in special techniques have been confined almost entirely to research laboratories. The development of the high intensity X-ray source and sensitive Geiger Counter Spectrometer offered the possibility of a simple rapid method of chemical analysis by secondary X-ray emission spectra without the use of gas X-ray tubes, vacuum spectrograph, photographic process, and microphotometer.

Experimental

As the Norelco Geiger Counter Spectrometer was developed for the powder diffraction technique, it was necessary to design and construct a special sample holder for secondary

emission work. The tube housing and X-ray tube were replaced by a sample holder carrying the regular collimating slit system. The sample was placed in a small depression milled in a thin sheet of lucite which was slipped into position on a slanting mount under the high intensity tube in the holder. A single crystal of calcite or sodium chloride placed with its 100 face parallel to the face of the regular sample holder was used for the diffraction grating. The AEG-50 tube was connected by a flexible shockproof cable to a Hayes diffraction unit power supply with variable controls to provide the maximum rating of the tube. An internal standard method of analysis, as used so effectively in spectrographic analysis, was found to give the best results. The intensities of the analysis and internal standard lines were determined by using the interval counter of the Geiger Counter Spectrometer.

Summary

A quantitative procedure was devised for the determination of RbCl in KCl with As_2O_3 as an internal standard, by using the K emission spectra. A working curve was prepared by plotting the percentage of RbCl against the ratio of intensity of the Rb Kα line to the intensity of the As Kα line. The range of concentrations of RbCl covered was from 0.05% to 5.00%, the amounts ordinarily encountered in minerals containing rubidium. The accuracy achieved was superior to the longer, more tedious chemical methods, and applicable to a lower limit of concentration. It is necessary for the samples used in preparation of the working curve to be similar in composition to the samples to be analyzed.

The method should find a wide field for application, especially in the analysis of materials that are extremely difficult to separate chemically, such as, the rare earths. In preliminary investigation, the analysis of rare earth elements using the L series emission lines, appeared very

promising, particularly for those above neodymium which are the more difficult ones to analyze chemically. Routine quantitative procedures for rare earths and thorium are being developed at present.

It is doubtful if the method can compete in the analysis of elements readily separated and analyzed by chemical means with a high degree of precision. However, it is believed that the limit of precision by the X-ray method is governed by the stability of the primary source. A better stabilizer or constant voltage source should improve the precision appreciably. Where time, rather than high precision, is the primary factor in analysis the X-ray method should compete even in some ordinary chemical analyses.

PART II

THE PREPARATION AND STUDY OF THE ANTIGEN FROM *EBERTHELLA TYPHOSA* KILLED BY X-RAYS

Introduction

The first suggested biological application of X-rays, following their discovery in November, 1895, was their effect on bacteria. An account of all the experiments on bacteria would occupy an entire volume. Studies of the effect of X-rays on bacteria in general and on *Eberthella typhosa* in particular have been reported by many investigators (7-12). Although there is conflicting evidence, in general, the results indicate that the effect of X-rays is independent of wave length and depends only on the amount of incident energy. Several workers have substantiated the fact that X-rays kill bacteria in a semilogarithmically linear fashion. Little work

has been done on the antigenic properties of bacteria killed by X-rays.

Experimental

The Machlett AEG-50 tube designed especially for such studies appeared to offer an excellent means for the preparation of sufficiently large quantities of antigen from X-ray killed bacteria for practical use. The tube was mounted in a lead covered box and was connected by a shockproof cable to a Hayes diffraction unit power supply. The dishes containing the bacteria in suspension were placed beneath the window of the tube for irradiation. Comparison of the antigen prepared by X-radiation with normal heat-killed antigen was made by immunizing rabbits to obtain blood sera from the two antigens. Reciprocal absorption experiments and cross titrations of the two antigens and sera were performed.

Summary

A method was developed using the high intensity Machlett AEG-50 X-ray tube to kill saline and aqueous suspensions of *Eberthella typhosa*. The cells killed by this method may act as an effective antigen. Preliminary work on suspensions of spore forms of *Bacillus Cereus* showed the method to be almost as effective in killing spore forms of bacteria as the normal forms. Special cells in which a thin film of the bacterial suspension is allowed to flow in close proximity to the window of the X-ray tube could be constructed for commercial production of antigen. It is demonstrated that the X-ray method not only competes favorably with the Oppenheimer ultraviolet irradiation technic which has a lethal effect in some cases in $\frac{1}{3}$ second, but has some marked advantages.

PART III

THE USE OF MONOCHROMATIC X-RAYS IN RADIOLOGY

Introduction

The use of X-rays in the medical field has grown to the extent that the subject is often regarded as a distinct science, radiology, consisting of two general divisions: diagnostic radiography and therapy. In a report in 1944 on the development of a monochromatic radiographic method for aluminum castings, Clark and Eyler (3) suggested that the use of a monochromatic X-ray source might be even more important in studying certain fine structures in medical radiography. Earlier, the superiority of monochromatic radiation was demonstrated by Clark and Gross (13). The extension of the use of monochromatic X-rays in medical diagnosis and therapy was undertaken in response to the many requests from the medical profession.

Experimental

Since the only radiographic X-ray tubes commercially available have tungsten targets, experiments with characteristic radiation from targets of lower atomic number were done with Machlett Type A-2 diffraction tubes operated in a Hayes unit. Shockproof tubes were clamped on a portable stand and connected by a flexible cable to the power supply. The diffraction tubes available had targets, respectively, of molybdenum, copper, cobalt, iron, and chromium. A Picker 150 KV Industrial Mobile X-ray Unit employing an oil-cooled tungsten target tube similar to medical tubes was used for comparison. Radiographs and phantom measurements were taken by standard procedures familiar to radiologists.

Summary

The use of monochromatic X-radiation in medical radiography and superficial therapy was investigated.

Radiographs taken with the characteristic radiation from molyddenum, copper, iron, cobalt, and chromium target diffraction tubes were superior in definition to those taken with conventional medical tungsten target tubes. Detailed technics for the skull using molybdenum radiation with both Parspeed and Fluorazure intensifying screens were developed and checked. The quality of the radiographs was superior and the time required was less than that required for tungsten radiation. It is believed that molybdenum radiation would be most generally useful in medical radiography because of its deeper penetration. However, the longer wave lengths of copper, cobalt, and iron target tubes might be very useful in the radiography of soft tissue and thin sections. The sharper definition obtained with monochromatic X-rays should prove to be especially useful in the almost undeveloped field of distortion radiography.

The use of monochromatic radiation for skin therapy was studied by determining the depth of penetration of the characteristic radiation into pressedwood blocks and by phantom measurements through water. Comparisons between molybdenum and tungsten radiation were made. Since the longer characteristic radiation of molybdenum is more readily absorbed in the surface layers, it should be particularly effective in superficial therapy. No phantom measurements on the longer wave length sources were made, but it is felt that the characteristic radiations of copper, cobalt, iron, and possibly chromium might be even more effective since the tube could be operated at a lower voltage to obtain the characteristic rays without exciting the more penetrating general radiation produced by higher voltages. A study was made of the deviation from the inverse square law in calculating dosages at different distances. Air

absorption of the longer wave lengths of iron and chromium makes it impossible to use the inverse square law in determining the dosages at distances not actually calibrated.

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VITA

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In 1938, he entered the Graduate School of the University of Chicago and was awarded the degree of Master of Science in Chemistry in 1940.

From 1940 to 1945, he was on the staff of the Geochemical Section of the Illinois State Geological Survey, Urbana, Illinois. During this time, he studied part-time in the Graduate School of the University of Illinois.

From September, 1945, to June, 1947, he served as a teaching assistant in the Division of Analytical Chemistry at the University of Illinois. He married Jean L. Rosselot on July 27, 1945.

He is a member of the following organizations: The American Chemical Society, Sigma Xi, Phi Lambda Upsilon, and Alpha Chi Sigma.

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"Effect of Oxidation on Reactivity and Swelling of Illinois Coals." *Ind. Eng. Chem.*, **35,** 346 (1943). (With O. W. Rees.)